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Title: **"Femtosecond Photonics: Fundamental Phenomena and Device Behavior"**

Principle Investigators: **E. P. Ippen, J. G. Fujimoto, and H. A. Haus**

Institution: Research Laboratory of Electronics
Massachusetts Institute of Technology
Cambridge, MA 02139

Summary:

This report describes a number of different research projects carried out under this contract. Emphasis is placed on progress made during the third and final year; but a listing of publications and reports covering all three years of the contract is attached. Titles of the individual projects described herein include:

Picosecond Optical Switching
Additive Pulse Modelocking
Ultrashort Pulse Generation in Titanium Sapphire
Additive Pulse Modelocking in Diode Pumped Nd:YAG and YLF
Multistage High Repetition Rate Femtosecond Amplifiers
Control of Spontaneous Emission with Semiconductor Microcavities
Carrier Dynamics in Metals and Semiconductors
Femtosecond Studies of Superconductors
Time Domain Interferometry
Nonlinear Dynamics in Active Semiconductor Devices
Four Wave Mixing and Information Storage in Photorefractive Crystals
Impulsive Excitation of Coherent Phonons

Also attached is a list of 29 journal publications, 4 book chapter contributions, and 35 conference presentations acknowledging this contract.

Final Report
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Principle Investigators: **E. P. Ippen, J. G. Fujimoto, and H. A. Haus**

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1. Picosecond Optical Switching

All optical switching with subpicosecond pulses has been pursued by our group for several years[1,2]. We have concentrated on interferometric switching using the index nonlinearity of optical materials. Lately we have concentrated on the construction of switches using fiber interferometers, because of the close to ideal behavior of optical fibers[2]. Our work is aimed at establishing a "proof of principle," exploring the requirements that must be met by the physical system in order to achieve satisfactory performance. Eventually, when quantum wells or other "engineered" materials of sufficient nonlinearity and of acceptably low linear and two-photon absorption will become available, then the principles demonstrated with the fiber system can be implemented in more practical systems with less "latency."

The fundamental requirement of a practical switch is that the output be a reasonable replica of the input. Nonlinear interaction using the Kerr effect (third order nonlinearity), by its nature, tends to distort the spectrum and pulse shape, the latter due to group velocity dispersion. A working switch must overcome this tendency of pulse distortion. One way to accomplish this is to use soliton-like interactions[3,4]. If this operating principle is chosen, the interaction region must possess negative dispersion, if the Kerr nonlinearity is positive, and viceversa. The "collisions" of the control pulses, and controlled pulses must be soliton collisions, so as not to distort the pulses, or soliton-like, if the system is not strictly a soliton system. Strict soliton collisions call for the use of different frequencies for the colliding pulses, sometimes an unacceptable constraint. If the colliding pulses have

the same frequency, they must be distinguishable by e.g. polarization. Two orthogonally polarized pulses do not interact in general in a distortion-free way. Distortion can be minimized, if the collision is "weak." In order to achieve large effects, the collisions must be repeated several times.

This operation principle has been chosen for a switch realized in our laboratory using a fiber ring reflector interferometer[5,6]. The collisions of orthogonally polarized pulses, traveling at different velocities due to fiber birefringence, were repeated by splitting the fiber into 11 segments, in each of which one collision occurred; the effects of the collisions were cumulative. The interaction was distortion-free as anticipated. The operating principle required the use of a polarization sensitive coupler. Because the coupler was not performing to specifications, the contrast ratio was not large. Yet the performance was in good agreement with theoretical predictions.

[1] A. Lattes, H. A. Haus, F. J. Leonberger, and E. P. Ippen, IEEE J. Quant. Electron. **QE-19**, 1718-1723 (1983).

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2. Additive Pulse Modelocking

Additive Pulse Modelocking (APM) is a novel scheme for the production of short pulses, particularly from solid state lasers with long gain-relaxation times[1]. These laser systems cannot be modelocked in a way analogous to the dye laser systems, in which the saturable gain and the saturable loss cooperate in the pulse shaping process.

Thus far, most APM systems employ a coupled cavity system, one cavity containing the laser medium, the auxiliary cavity containing a Kerr medium, generally a fiber. The

length of the auxiliary cavity needs to be stabilized by a feedback circuit to maintain the relative phase of the pulses meeting at the coupling mirror between the two cavities.

We have developed the criteria for self-starting of the APM modelocking process[2]. When the process is self starting, the need for an internal modulator is obviated, not only simplifying the system, but also eliminating the competition between the modulator frequency and the pulse repetition frequency. It is this competition that can render the mode locking process unstable.

We have APM modelocked a flashlamp pumped Nd:YAG laser achieving 6 ps pulses without sacrifice of average power[3]. A diode laser pumped Nd:YAG crystal gave 2 ps modelocked pulses[4]. We have achieved APM action in a Ti:Sapphire laser in a single cavity, analogous to the system demonstrated first by Sibbett et al. It is generally necessary to start this system with a moving mirror in an external cavity.

These experimental results have stimulated theoretical work. The APM principle applies to any interferometric transformation of nonlinear phase modulation to nonlinear amplitude modulation. The single cavity Ti:Sapphire system operates in this way, the role of the two arms of an interferometer being played by two transverse cavity modes. A theory that considers many possible configurations that produce APM action is currently under investigation.

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[2] E. P. Ippen, L. Y. Liu, and H. A. Haus, "Self-starting condition for additive-pulse mode-locked lasers," Opt. Lett. 15, 183-185 (1990).

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3. Ultrashort Pulse Generation in Titanium Sapphire

The Ti:Al_2O_3 laser is an important model system for investigating ultrashort pulse generation in solid state lasers. The properties of Ti:Al_2O_3 are expecially attractive for ultrafast spectroscopy. Ti:Al_2O_3 features a tuning range from 700 nm to 1100 nm with room temperature operation and high thermal conductivity ¡and high energy storage[1]. The broad gain bandwidth of this material makes it ideal crystal for the generation and amplification of femtosecond pulses. The tuning range is especially suited for studies of GaAs and AlGaAs-based opto-electronic devices. In addition, amplification and frequency conversion techniques may be developed to produce tunable ultraviolet pulses for femtosetcond UV spectroscopy. For these reasons, the investigation of ultrashort pulse generation in Ti:Al_2O_3 has recently emerged as an active and promising area of research.

Working in collaboration with Dr. P.A. Schulz of M.I.T. Lincoln Laboratory we have recently developed the APM technique for ultrashort pulse generation' in Ti:Al_2O_3[2]. Additive Pulse Modelocking in Ti:Al_2O_3 is significant because it was the first demonstration of self starting passive modelocking without the need for active gain or loss modulation. Short pulses can be generated with a significant reduction in cost and complexity over previous approaches.

The APM laser generates short pulses using an external cavity containing a Kerr medium (a single mode optical fiber of appropriate length) which has an intensity dependent index of refraction. The external cavity functions as a nonlinear Fabry Perot with an intensity dependent reflectivity. If the external cavity length is interferometrically controlled relative to the main cavity, it is possible to operate the external cavity as a fast saturable absorber. Pulses as short as 1.4 ps have been generated directly from the Ti:Al_2O_3 laser. Using an intracavity prism pair with negative group velocity dispersion to remove pulse chirp and produce pulse compression resulted in bandwidth limited pulses of 230 fs[3]. Pulses of similar duration can also be achieved by external dispersion compensation by a diffraction grating pair[4].

During the last year our research has focussed on understanding the starting dynamics of the APM modelocking[5]. Studies of starting dynamics provide an approach for investigating the mechanisms of the pulse formation process. Our investigations demonstrate that the nonlinear external cavity produces pulse shaping by a fast saturable absorber like action. These studies provide important design criteria for optimizing the laser system as well as for generalizing the APM technique to other solid state laser materials.

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4. Additive Pulse Modelocking in Diode Pumped Nd:YAG and YLF

We have also demonstrated self-starting APM in diode-pumped Nd:YAG and Nd:YLF[1,2]. The diode pumped Nd materials are especially attractive since they can be engineered into a compact and low cost ultrashort pulsed laser technology.

In our experiments three diode arrays were used as for pumping. The main laser cavity consisted of a high reflector, a folding mirror, and an output coupler. The external cavity consisted of a beam splitter, an optical fiber, and a retroreflecting mirror. The cavity length was 1.1 m, corresponding to a 136 MHz repetition rate. In Nd:YAG durations of 1.7 ps were obtained with a spectral bandwidth of 0.67 nm[2]. These are the shortest pulses produced to date directly from an Nd:YAG laser. In Nd:YLF, chirped pulses of 2.0 ps with a bandwidth of 0.8 nm were generated[1].

These results demonstrate that can be scaled to lower power systems such as diode pumped solid state lasers. Pulse durations are generated which are significantly shorter than possible by previous techniques. Finally, diode pumped solid state lasers can be engineered into a compact and low cost ultrashort pulse technology.

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[2] J. Goodberlet, J. Jacobson, J. G. Fujimoto, P. A. Schulz, and T. Y. Fan, "Self starting additive pulse modelocked diode pumped Nd:YAG laser," Opt. Lett. 15, 504-506 (1990)

5. Multistage High Repetition Rate Femtosecond Amplifiers

Dye laser systems and flowing dye amplifiers are currently the most widely used technology for ultrashort optical pulse generation. We are continuing our research on dye based systems in order to enhance our experimental facilities for investigating ultrafast phenomena. We have recently completed the development of a multistage, high repetition rate, dye amplifier which may be used for a variety of ultrafast studies in materials and devices.

Our femtosecond pulse laser system is based on a colliding-pulse modelocked ring dye laser (CPM)[1]. The CPM generates 35 fs pulses at a wavelength of 630 nm. The advantage of the CPM laser is that it produces extremely short pulse durations. However, since the CPM uses a passive modelocking with saturable absorber dyes, the output is not tunable in wavelength. This trade off between short pulse duration and wavelength tunability is typical of ultrafast laser systems, and much of our work focuses on the development of new ultrafast generation techniques to achieve tunable sources.

In order to generate high intensities necessary for studies of nonlinear proceses or frequency conversion and pulse compression, the femtosecond pulses generated by our CPM are amplified by a copper vapor laser pumped dye amplifier[2]. The copper vapor laser amplifier operates at 8 kHz repetition rate. The high repetition rate permits the use of lock-in detection and signal averaging to achieve high sensitivity experimental measurements. We have recently completed the development of a novel multistage copper vapor laser pumped amplifier system[3].

The amplifier system has been designed with modular construction and in a flexible arrangement so it may be configured for amplification, white light continuum generation, or ultrashort pulse compression. The system femtosecond pulses with 20 - 30 μJ pulse energy with pulse durations of 50 fs corresponding to peak intensities in excess of 100 MW.

When an intense ultrashort optical pulse is focussed into a material with an intensity dependent index of refraction, self phase modulation effects can be used to broaden the spectrum of the pulse. In the high intensity limit, the spectral broadening becomes very pronounced and a broadband white light continuum is generated with wavelengths ranging from 400 nm to greater than 900 nm[4]. The technique thus provides a source of tunable femtosecond light for experimental studies.

Although continuum generation has been widely used experimentally, the physical origins of the process are not well understood. We are currently investigating the nonlinear frequency modulation and beam propagation effects associated with high peak intensity pulses. These investigations are important because they suggest other technques for nonlinear frequency generation. In preliminary work, we have observed that the continuum is generated coherently, and by using negative group velocity dispersion, it is possible to compress selected wavelength regions of the continuum to less than 20 fs. This represents a powerful new capability for ultrafast spectroscopy.

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6. Control of Spontaneous Emission with Semiconductor Microcavities

Optical microcavities hold technological promise for constructing efficient, high speed semiconductor lasers. One particularly interesting possibility is the alteration of the spontaneous emission rate of the device by the presence of the cavity. Such alteration has previously been observed with atoms but is more difficult to achieve in a semiconductor device because the broad spontaneous emission bandwidth requires cavity dimensions on the order of a wavelength. To determine the potential feasibility and significance of spontaneous emission alteration in these devices, we have analyzed the radiation modes of oscillating dipoles in planar (one dimensional confinement and optical-wire (two dimensional confinement) structures[1]. We find that an idealized planar metallic mirror cavity can suppress the spontaneous emission by no more than a factor of two with respect to free space. The amount of suppression obtainable with a real dielectric stack will be even

less. Theory shows, however, that much larger effects could be achieved by restricting the dimensionality to that of the optical wire. Enhancement of spontaneous emission should be more easily observable, and this has been shown to be the case[2]. With GaAs quantum-wells, monlithically integrated with Fabry-Perot cavities fabricated at NEC, we have observed enhancement of emission by a factor of two and a corresponding reduction in the luminescence lifetime due to cavity effect. Future work on this topic will rely on advances in the fabrication of suitable wire or dot devices or of improved resonator structures.

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7. Carrier dynamics in Metals and Semiconductors

Working in collaboration with researchers at the General Motors Research Laboratories, we have investigated the dynamics of image potential states in metals[1-3]. An image potential state occurs in a metal when an electron outside the surface of the metal is bound state to its image charge in the bulk. Electrons in the image potential state form a Rydberg series and are a two dimensional electron gas analogous to quantum well systems in semiconductors. The electrons relax by tunnelling from the image potential state back to the bulk states. The investigation of image potential states is thus an important approach to understanding ultrafast electron dynamics in metals.

In order to study femtosecond image potential dynamics, we have developed new measurement techniques which combine photoemission spectroscopy with femtosecond optics. An ultrashort pump pulse is used to prepare the excited state while a delayed pump pulse is used to photoionize the state. The photoemitted electrons are energy analyzed as function of delay between the pump and probe pulses. This permits a transient measurement of photoemission spectra on the time scale of 10 fs.

Using these techniques, we have performed a comprehensive investigation of the image potential states in Ag. These studies are of interest because they permit a test of theoretical predictions of image potential dynamics. Relaxation dynamics of the $n = 1$ and $n = 2$ states on the 100 and 111 surfaces were studied. The dynamics of the image potential state have been measured as a function of time and electron energy. The lifetime of the $n = 1$ state on Ag(100) was 25 ± 10 fs. To our knowledge, this measurement represents the

highest time resolution photoemission measurement to date[2]. Systematic measurements of lifetimes of different states in the Rydberg series on different surfaces have been performed and compared to theoretical descriptions of the image potential dyanmics based on tunneling and many particle models[3].

We have also continued our work on femtosecond carrier dynamics in semiconductors. We have established a collaborative program with condensed matter theorists from the University of Florida[4]. Our objective is to combine state of the art experimental and theoretical techniques to investigate fundamental excited carrier dynamics in technologically relevant compound semiconductors and quantum confined structures. Within this collaborative program we have begun to develop a comprehensive model for carrier dynamics in the GaAs and AlCaAs semiconductors. This will result in a powerful tool for the prediction of nonequilibrium behavior in a variety of new materials.

Research at M.I.T. focusses on femtosecond experimental studies in GaAs and AlGaAs, while our collaborators at the University of Florida perform theoretical investigations of carrier dynamics using full band structure and ensemble Monte Carlo techniques. The Monte Carlo simulation is used to find the electron and hole distribution functions by developing a correspondence with experimentally measured differential transmission pump probe data. These studies show that it is essential to include collisional broadening during photoexcitation and the effects of hole scattering in the theoretical model[5,6]. The combination of theoretical and experiment studies provided the first direct evidence for hole redistribution on a femtosecond time scale.

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[2] R. W. Schoenlein, J. G. Fujimoto, G. L. Eesley, and T. W. Capehart, "Femtosecond dynamics of the $n = 2$ image-potential state on Ag(100)," Phys. Rev. B 41, 5436 (1990).

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8. Femtosecond Studies of Superconductors

When an ultrashort optical pulse is incident on the surface of a metal, most of its energy absorbed directly, because of the high electron density, into the free electron gas. The resulting rise in electron temperature produces a dynamic change in reflectivity. Relaxation of this change occurs as the electrons lose energy to the lattice via phonon emission. The rate is governed by the electron-phonon coupling strength. Since the strength of the electron-phonon coupling is an important component in the BCS theory of superconductivity, we were motivated to undertake a systematic study of these dynamics in superconductors. This was done in collaboration with Prof. M. Dresselhaus' group. In a series of experiments[1] we measured λ the relaxation rate, for ten different metals (4 superconducting and 6 not). The agreement between the values obtained and those derived from the literature is strikingly good. Apparent advantages of our method over other techniques (e.g. tunneling or heat capacity measurements) for measuring λ are that it is a direct measurement, it works at room temperature, and it can be applied to non-superconducting as well as superconducting samples. In some metals for which the changes in reflectivity were otherwise too small to detect, we have also found that thin overlayers of Cu (which has *d*-band transitions in the visible) can be used to enhance greatly the experimental reflectivity changes without affecting the inherent relaxation rate. This extends the method to virtually any metal film.

Emboldened by the success of these results we also performed several preliminary pump-probe reflection and transmission experiments on three high T_c thin films: $YBa_2Cu_3O_{7-x}$, $Bi_2Sr_2CaCu_2O_{8+x}$ and $Bi_2Sr_2Ca_2Cu_3O_{10+y}$[2]. For these materials, of course, we do not yet have a theoretical framework with which to connect our experiments to high T_c superconductivity. Nevertheless, in these preliminary experiments, we have observed strong changes in observed relaxation rates with changing T_c.

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[2] S. D. Brorson, A. Kazeroonian, D.W. Face, T.K. Cheng, G. L. Doll, M.S. Dresselhaus, G. Dresselhaus, E.P. Ippen, T, Venkatesan, X. D. Wu, and A. Inam, "Femtosec-

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9. Time Domain Interferometry

Investigations of nonresonant nonlinear process in semiconductors are directly relevant to the development of high-speed all-optical switching devices and the optimization of high speed modulation performance in diode lasers. In particular, the characterization of the nonlinear index of refraction, n_2, and its dynamics is key to the development of such fast devices.

Our group has recently developed a novel technique for performing highly sensitive nonlinear index measurements[1]. This technique is called time division interferometry or TDI and uses a single waveguide with time division multiplexing to perform transient pump probe interferometric measurements of n_2. A pump and time delayed probe pulse are coupled into a waveguide structure. The transient phase shift of the probe pulse produced by the pump is measured by interfering the probe with a time division multiplexed reference pulse. The femtosecond transient behavior of the nonlinear index can be measured by varying the delay between the pump and probe pulses. The TDI technique reduces parasitic contributions from thermal and acoustic effects and achieves a measurement sensitivity of $\lambda/500$ without active length stabilization of the interferometer. Active stabilization increases the sensitivity by over an order of magnitude. Using this technique we have performed the first direct measurements of the nonresonant nonlinear index of AlGaAs[2].

Nonlinear effects other than the nonlinear index of refraction n_2 can also be studied using time domain techniques. The nonlinear two photon absorption β is an important limiting process for all-optical switching since it produces excited carriers which limit the recover times of the index nonlinearity. We have performed measurements of β and the associated carrier dynamics for AlGaAs waveguide devices[3]. Coupled with measurements of linear properties and nonlinear index, this constitutes a complete characterization of the waveguide device[4]. This information can be used to calculate the all optical switching behavior and determine figures of merit for all optical switching.

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[2] M. J. LaGasse, K. K. Anderson, C. A. Wang, H. A. Haus, and J. G. Fujimoto, "Femtosecond measurements of the nonresonant nonlinear index in AlGaAs," Appl. Phys. Lett. 56, 417 (1990).

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[4] K. K. Anderson, J. J. LaGasse, H. A. Haus, and J. G. Fujimoto, "Femtosecond time domain techniques for charactertization of linear and nonlinear optical properties in GaAs waveguides," Mat. Res. Soc. Symp. Proc. 167, 51 (1990).

10. Nonlinear Dynamics in Active Semiconductor Devices

Nonlinear optical effects in active waveguides not only influence the generation and propagation of ultrashort pulses in diode lasers they may also find application in all-optical switching. In our laboratory, with 100 fs-duration pulses in the 800-900 nm regime (obtained by fiber compression of synch-pumped dye laser pulses) and with similar pulses in the 1.45-1.65 μm band (from an APM F-center laser), we have been able to perform the first investigations of nonlinear dynamic behavior in both GaAlAs[1] and InGaAsP[2] devices under various excitation conditions. By varying the wavelength of the pump and probe beams, as well as injection current in our diode structures, we have studied interactions in the presence of gain, loss, or nonlinear transparency. In all cases there is an injected carrier density on the order of $10^{18}/cm^3$, and this makes the nonlinear optical behavior considerably different from what is observed in passive devices or pure materials. In both GaAlAs and InGaAsP devices we have discovered a strong nonlinearity due to nonequilibrium between the carrier and lattice temperatures. Heating of the carrier gas with respect to the lattice has a recovery time on the order of 1 ps in GaAlAs and 650 fs in InGaAsP; and, since heating occurs via free electron absorption and no change in carrier number is involved, recovery is complete. This is a particularly important characteristic for all-optical switching applications. Our most recent experiments have yielded preliminary measurements of femtosecond index of refraction dynamics as well as gain changes In GaAlAs. Index changes corresponding to optical Kerr effect and nonequilibrium heating have been observed and are comparable in magnitude to those produced by population changes. During the past year we have also used a novel means for detecting these nonlinear optical interactions by monitoring changes in diode voltage[3]. By measuring bias voltage as a function of time delay between two optical pulses passing through the diode, we can clearly identify nonlinear optical interactions that utilize active carriers. The time constants observed corroborate those obtained from pump-probe measurements of nonlinear gain.

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11. Four Wave Mixing and Information Storage in Photorefractive Crystals

Photorefractive materials such as $BaTiO_3$, SBN, and $LiNbO_3$ present large optical nonlinearities that are attractive for applications in optical devices based on four-wave mixing processes[1]. Although the response times of these crystals are typically in the millisecond range, they provide an important model system for the design of phase conjugation, optical processing, and optical logic techniques. Working in collaboration with investigators from Tufts University, we have performed the first four-wave mixing experiments with $BaTiO_3$ using femtosecond optical pulses[2].

These investigations explore the factors which determine the temporal broadening of optical signals in four wave mixing. Studies were performed using 40 fs pulse durations from a CPM dye laser. Different phase conjugation geometries were examined including the ring resonator as well as the two beam coupling geometry. A surprising finding was that temporal signals are influenced only by material dispersion effects and that pulse durations of 40 fs could be preserved in the four wave mixing process. Since four wave mixing in $BaTiO_3$ occurs via the photorefractive effect, these studies determine the transient behavior of scattering from volume index photorefractive gratings.

Four wave mixing in $BaTiO_3$ is a well established approach for encoding image and phase conjugation information. We have extended these concepts and demonstrated the encoding of temporal information using a two beam four wave mixing approach. Our experiments are closely related to femtosecond holography which uses holographic recording to store transient femtosecond images. In our approach however, the temporal behavior of a signal pulse can be encoded geometrically onto the volume photorefractive grating which is written in the $BaTiO_3$ crystal. This temporal signal can subsequently be read out by diffracting as probe pulse from the volume grating. These investigations suggest a new approach for encoding and reconstructing high speed optical information. Extensions of these techniques using acousto-optic modulators or other programmable volume diffraction devices may make it possible to generate programmable optical pulse trains at THz repetition rates.

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12. Impulsive Excitation of Coherent Phonons

We have recently reported the first observations of coherent optical phonon excitation in two opaque conducting materials, bismuth and antimony[1]. Previous experiments involving excitation of coherent phonons in transparent materials have relied upon stimulated Raman scattering as the excitation mechanism and have utilized changes in transmission for detection. In our work we simply observe changes in sample reflectivity following absorption of a femtosecond pulse incident upon the surface. The reflectivity is observed to oscillate at the frequency corresponding to the A_{1g} mode in each case (2.9 THz in Bi and 4.5 THz in Sb) indicating that the modulation varies linearly with phonon amplitude. Both the large amplitudes of the reflectivity changes (greater than 10^{-3}) and the absence of other allowed Raman modes argue that a mechanism other that stimulated Raman scattering is the driving force. The initial phase of the oscillations (cosinusoidal rather than sinusoidal) also imply that an electronic transition is involved. Experiments are in progress to clarify the actual mechanism and to use this technique to study electron-phonon interactions. The method opens up the possibility for detailed time-domain studies of phonon dynamics on a whole class of opaque materials.

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